# ISOTOPES

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# PREFACE

I HAVE undertaken the preparation of this book on Isotopes in response to many requests made to me by teachers of physics and chemistry and others working in these subjects that I should publish the results obtained by means of the Mass-spectrograph in a form more convenient to the public than that in which they first appeared. This is one of the reasons why the space allotted to the inactive isotopes may appear, in the light of the general title of the book, somewhat disproportionately large. Another is that the subject of radioactive isotopes really requires a book to itself, and I am in the hope that the inadequacy of my account may stimulate the production of such a volume by hands more competent than mine to deal with this very special and remarkable field of modern science. The logical order of exposition of a scientific subject is to start with the simple and from that build up the more complex. Unfortunately the sequence of events in experimental research is the exact opposite of this so that a compromise must be effected, unless one is content to sacrifice historical treatment altogether. The latter seems very undesirable in a new subject. I have endeavoured in Chapters I, II and IV, and elsewhere when possible, to adhere strictly to the historical order of events even at the cost of some reiteration.

I wish to take this opportunity of expressing my indebtedness to Mr. C. G. Darwin for his timely criticism and unfailing assistance throughout the work, and also to Mr. R. H. Fowler for help with the proofs. My thanks are also due to Professor Soddy for his diagram of the radioactive isotopes, to Mr. A. J. Dempster for kindly sending me the illustrations of his work,

to the proprietors of the *Philosophical Magazine* and to
the Council of the Chemical Society for permission to use the
plates and figures of my original papers, and to Messrs.
Macmillan & Co., for the diagram of the radioactive trans-
formations.

<div align="right">F. W. ASTON.</div>

CAMBRIDGE,
    *January*, 1922.

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# CHAPTER I

## INTRODUCTION

**1. Introduction.**—Towards the end of the last century the attitude of science in relation to the atomic theory started to undergo a complete and radical change. What had been before regarded as a convenient working hypothesis became with remarkable rapidity a definite statement of fact.

This transformation is now complete and in any well-equipped laboratory to-day not only can individual atoms be detected but the movements of the swiftest of them can be tracked and made visible even to the untrained eye.

The causes of this remarkable advance are to be ascribed in particular to the discovery of radioactivity, which has provided us with atomic projectiles possessing enough energy to produce visible and measurable effects individually, and in general to the steady and continuous improvement in technical methods. Subject to such unprecedented scrutiny it was to be expected that the fundamental physical theories which underlie the applied science of chemistry and form a solid mathematical foundation for its formulæ, might show hitherto unsuspected flaws. Such expectations began to be realised when, among the radioactive elements, Boltwood failed to separate ionium from thorium, and, among the inactive elements, when Sir J. J. Thomson a few years later observed the anomalous behaviour of neon when subjected to positive ray analysis. Further and still more delicate and careful scrutiny of these flaws revealed them, as it must always do, if they are real, not as fortuitous and disconnected but as a definite and ultimately intelligible pattern. It is with the interpretation of this pattern, so revealed, that this volume is concerned, so that it will be of interest to look back rather over a century to the

beginning of the theories which form the background against which it was first observed.

**2. Hypotheses of Dalton and Prout.**—In the generalisation, known as the Atomic Theory, put forward by Dalton in 1803, which laid the foundations of the whole of modern chemistry, five postulates were laid down, and it is a striking tribute to the shrewd intuition of that observer that, of those five, to this day, the validity of one only is in any question. This postulate is that :—Atoms of the same element are similar to one another and equal in weight. It obviously consists of two parts and if we combine both as a definition of the word element the whole becomes a truism ; this aspect of the matter will be considered later on. For the present we shall take the word "element" to mean what Dalton evidently intended it to mean, and what we generally consider it to mean to-day, namely a substance such as chlorine or lead which has constant chemical properties, and which cannot be resolved into further components by any known chemical process. The first half —taken together with the other four postulates—is then sufficient to define the word "element" and the second becomes a pure hypothesis.

About ten years later Prout suggested that the atoms of the elements were all made up of aggregations of atoms of hydrogen. On this view the weights of all atoms must be expressed as whole numbers, and if, as postulated by Dalton, the atoms of any particular element are all identical in weight, the atomic weights and combining ratios of all elements must be whole numbers also. Chemists soon found that in the case of many elements this was certainly not in agreement with experiment ; the more results they obtained the more impossible it was to express the atomic weights of *all* the elements as whole numbers. They therefore had to decide which hypothesis, Dalton's or Prout's, they would adopt. There was little doubt as to the result of the decision and in due course Prout's theory was abandoned.

It is interesting to consider the reasons which led to a decision which the subsequent history of science proves to have been as wise in principle as it was wrong in fact. The alternative

views were—either an element was composed of atoms of identically the same weight, when certain elements the weights of the individual atoms must be fractional. or these particular elements were composed of atoms of different weights mixed together, so that though the individual weights of the atoms would still be whole numbers their mean would be a fraction. It is almost inconceivable that the second alternative never occurred to philosophers during the time when the decision hung in the balance—indeed it was far more likely to be considered then than years later when Dalton's view had been generally accepted—but the objections to it were immediate and formidable. The idea that particles could behave in a practically identical manner even though they had different weights is not one that commends itself, *a priori*, to common sense, and as a working hypothesis for chemists it is as hopeless and indefinite as the simpler alternative is distinct and inspiring. Also it could be urged that the objections to the fractional weights of atoms were rather philosophic than practical. They were concerned with the structure of individual atoms and so might be, and wisely were, set aside till the time, distant enough it would then have seemed. when these hypothetical entities could be dealt with experimentally.

The idea that atoms of the same element are all identical in weight could not be challenged by chemical methods, for the atoms are by definition chemically identical and numerical ratios were only to be obtained in such methods by the use of quantities of the element containing countless myriads of atoms. At the same time it is rather surprising, when we consider the complete absence of positive evidence in its support that no theoretical doubts were publicly expressed until late in the nineteenth century, first by Schutzenberger and then by Crookes, and that these doubts have been regarded, even up to the last few years, as speculative in the highest degree. In order to dismiss the idea that the atoms of such a familiar element as chlorine might not all be of the same weight, one had only to mention diffusion experiments and the constancy of chemical equivalents. It is only within the last few years that the lamentable weakness of such arguments has been exposed and it has been realised that the experimental

separation of atoms differing from each other by as much as 10 per cent. in weight, is really an excessively difficult operation.

**3. Crookes' meta-elements.**—The chemist who above all others urged the possibility of the heterogeneity of atoms was the late Sir William Crookes, to whom we are indebted for so many remarkable scientific prophecies. His address to the Chemical Section of the British Association at Birmingham in 1886[1] is a most amazing effort of reason and imagination combined and should be read by all those interested in the history of scientific thought. In it he says : " I conceive, therefore, that when we say the atomic weight of, for instance, calcium is 40, we really express the fact that, while the majority of calcium atoms have an actual atomic weight of 40, there are not a few which are represented by 39 or 41, a less number by 38 or 42, and so on. We are here reminded of Newton's ' old worn particles.'

" Is it not possible, or even feasible, that these heavier and lighter atoms may have been in some cases subsequently sorted out by a process resembling chemical fractionation ? This sorting out may have taken place in part while atomic matter was condensing from the primal state of intense ignition, but also it may have been partly effected in geological ages by successive solutions and reprecipitations of the various earths.

" This may seem an audacious speculation, but I do not think it beyond the power of chemistry to test its feasibility."

Later [2] he developed this idea in connection with his pioneer work on the rare earths. By a laborious process of fractional precipitation he subdivided the earth yttria into a number of components which had different phosphorescent spectra but resembled each other very closely in their chemical properties. Pointing out that at that time yttrium was considered to be an element he says : " Here, then, is a so-called element whose spectrum does not emanate equally from all its atoms ; but some atoms furnish some, other atoms others, of the lines and bands of the compound spectrum of the element. Hence the atoms of this element differ probably in weight, and certainly

[1] *Nature*, **34,** 423, 1886.
[2] *Trans. Chem. Soc.*, **53,** 487, 1888 ; **55,** 257, 1889.

in the internal motions they undergo." He called such components "Meta-elements" and suggested that the idea might apply to the elements generally, for example referring to the seven series of bands in the absorption spectrum of iodine, "some of these molecules may emit some of the series, others others, and in the jumble of all these molecules, to which is given the name 'iodine vapour,' the whole seven series are contributors."

In so far as they differed a little in atomic weight and a mixture of them constituted a chemical element, these hypothetical meta-elements may be said to have offered the first feasible explanation of the fractional atomic weights. But as more and more refined chemical methods were applied, the rare earths one after another yielded to analysis and the different spectra observed by Crookes were shown to be due to the fact that he was dealing with a mixture of real elements, each of which had a characteristic spectrum and a definite atomic weight. The theory of meta-elements was therefore abandoned and the problem of fractional atomic weight remained unsolved.

**4. The discovery of Isotopes.**—As time went on the numbers representing the atomic weights grew more and more accurate and consistent. Significant figures one after another were added by one worker, confirmed by others, and finally approved by an International Committee. Small blame to the student therefore if, when studying the imposing list of numbers called the International Atomic Weights, he fell into the very natural error of confusing "atomic weights" with "weights of atoms," and considered that these figures did actually represent the relative weights of the individual atoms themselves.

Why so many of the atomic weights should be very nearly integers when expressed on the scale $O = 16$ was still a very difficult question to answer, for the probability against this being due to pure chance was enormous, but it was not until the discovery of radioactivity that the true reason for this curious jumble of whole numbers and fractions was suggested, and later confirmed generally by positive ray analysis. It is

worth noting that the first experimental proof that the atoms of an element might be even approximately of the same weight was given by positive ray parabolas.[1]

The results given by the radioactive elements introduced a wealth of new and revolutionary ideas. One of these was that elements might exist which were chemically identical but yet differed in radioactive properties and even in atomic weight. By 1910 this idea had gained ground and was seriously put forward and discussed by Soddy. At about the same period the technique of positive ray analysis was rapidly being improved, and in 1912 the first results were obtained from neon which were later to support this new idea and carry it into the region of the non-radioactive elements. From this time onwards advances were made in the two fields side by side, and so it happened that at the meeting of the British Association in 1913 [2] papers were read in different sections, one on the Radio-elements and the Periodic Law, the other on the Homogeneity of Neon, both of which tended to prove that substances could exist with identical, or practically identical, chemical and spectroscopic properties but different atomic weights.

The need for a specific name for such substances soon became imperative and Soddy suggested the word Isotopes (ἴσος equal, τόπος, place) because they occupied the same place in the periodic table of the elements.

[1] *V.* p. 29.
[2] Oddly enough this was the first meeting of the Association at Birmingham since the one twenty-seven years before at which Crookes made his prophetic remarks about atomic weights already quoted.

# CHAPTER II

## THE RADIOACTIVE ISOTOPES

**5. Chemical identities among the radioactive elements.**—Apart from the purely speculative considerations which have already been detailed, the theory of isotopes had its birth in the gigantic forward wave of human knowledge inaugurated by the discovery of radioactivity. It can admittedly be argued that, even if no radioactive elements existed, isotopes would inevitably have been discovered by the method of positive rays. But progress must then have been exceedingly slow, and the arrival at the real interpretation of the idea, depending as it does on Sir Ernest Rutherford's theory of the " nucleus " atom, almost impossible.

In 1906 Boltwood at Yale discovered a new element in the radioactive group which he called Ionium,[1] and described as having chemical properties similar to those of thorium. So much was this the case that if, by accident, salts of these two elements were mixed, he found it impossible to separate them again by any of the chemical processes.

Boltwood, being occupied in the experimental proof that ionium was the parent substance of radium, did not pursue this line of investigation further at the time, but the work was later taken in hand by Marckwald and Keetman of Berlin.[2] Thanks to the rapid advance in radioactive methods there were now at command means of detecting change in concentration of a delicacy unheard of in the previous work on the rare earths, but yet, after years of patient and laborious work, not the slightest sign of separation of ionium and thorium could be observed. The chemical similarity between these

[1] Boltwood, *Amer. J. Sci.*, **22**, 537, 1906 ; **24**, 370, 1907.
[2] Keetman, *Jahr. Radioactivitat*, **6**, 269, 1909.

two bodies was therefore of an order entirely different to that exhibited by the rare earth elements, and came as near absolute identity as the most critical mind could require.

This result was confirmed in the most rigorous manner by Auer v. Welsbach,[1] who was able to apply to the problem his valuable experience in work on the rare earths.

Furthermore, Mesothorium, discovered by Hahn in 1907, was shown to be chemically inseparable from radium by Marckwald [2] and Soddy [3] and similar chemical identities were shown to be exceedingly probable in many other cases of radioactive products. Certain regularities in the occurrence of these were pointed out by Hahn and Meitner.[4]

The situation was admirably summed up by Soddy in his report on radioactivity for the year 1910 [5] in the following words :—

" These regularities may prove to be the beginning of some embracing generalisation, which will throw light, not only on radioactive processes, but on elements in general and the Periodic Law. Of course, the evidence of chemical identity is not of equal weight for all the preceding cases, but the complete identity of ionium, thorium and radiothorium, of radium and mesothorium 1, of lead and radium D, may be considered thoroughly established. . . . The recognition that elements of different atomic weights may possess identical properties seems destined to have its most important application in the region of inactive elements, where the absence of a second radioactive nature makes it impossible for chemical identity to be individually detected. Chemical homogeneity is no longer a guarantee that any supposed element is not a mixture of several of different atomic weights, or that any atomic weight is not merely a mean number. The constancy of atomic weight, whatever the source of the material, is not a complete proof of homogeneity, for, as in the radioelements, genetic relationships might have resulted in an initial constancy of proportion between the several individuals, which no sub-

[1] A. von Welsbach, *Wien. Ber.* iia, **119**, 1011,   1910.

[2] Marckwald, *Ber. d. Chem. Ges.*, **40**, 3420,   1910.

[3] Soddy, *Trans. Chem. Soc.*, **99**, 72,   1911.

[4] Hahn and Meitner, *Physikal. Zeitsch.*, **11**, 493,   1910.

[5] Soddy, *Chem. Soc. Ann. Rep.*, 285,   1910.

sequent natural or artificial chemical process would be able to disturb. If this is the case, the absence of simple numerical relationships between the atomic weights becomes a matter of course rather than one of surprise."

**6. Spectroscopic identity of isotopes.**—The next great advance was of an even more revolutionary character. This consisted in the demonstration that the chemically indistinguishable products of the transformation of the radioactive elements might also be spectroscopically identical. The idea that elements of different atomic weight might yet have the same spectrum originated in Sir Ernest Rutherford's laboratory and appears to have been first entertained by A. S. Russell. With Rossi [1] he undertook the comparison between the spectrum of pure thorium and that of a mixture of thorium and ionium which radioactive evidence showed to contain a large percentage of the latter element. No new lines attributable to ionium were observed; in fact the spectra obtained were absolutely indistinguishable.

After giving in full the radioactive evidence as to the probable percentage of ionium present, and showing that it was practically impossible for this to be too small for its spectrum to appear, the writers go on as follows :—

" There are, however, two other possible ways of explaining our failure to obtain a distinct spectrum for ionium, besides the one discussed above. It is possible that :—

" (1) Ionium has no arc spectrum in the region investigated, or

" (2) Ionium and thorium have identical spectra in the region investigated.

" The first possibility is highly improbable, for all solids of high atomic weights have arc spectra, and, further, all rare earths have highly complicated spectra.

" The second possibility, though somewhat speculative in nature, is suggested by some recent work on the chemical properties of the radio-elements. There is no evidence at present to disprove its truth. It is well known that there are no less than four sets of longlived radio-elements, the

[1] Russell and Rossi, *Proc. Roy. Soc.*, **77A**, 478, 1912.

members of each of which are chemically non-separable. These elements do not all belong to the group of rare earths, many non-radioactive members of which are known to be chemically very similar. Mesothorium, for instance, which is chemically non-separable from radium, belongs to the alkaline earth group. Again the two non-separable $a$ ray products which are present in ordinary uranium, and which have been called by Geiger and Nuttall uranium I and uranium II belong to the chromium-molybdenum-tungsten group of elements. The explanation of these striking chemical similarities is very probably that the two very similar bodies are really different members of the same group of elements, the difference in their chemical properties being less pronounced than the difference between other members of the same group, owing to the small difference in their atomic weights. *But the possibility that they are identical in all physical and chemical properties, and differ only in atomic weight and in radioactive properties, should not be lost sight of. If this explanation should eventually prove justified, the spectrum of ionium would be identical with that of thorium.*"

It is not surprising that the idea was put forward with some caution. Unlike that of chemical identity which had been led up to by a gradual series of steps, it was entirely new and contrary to all the preconceived ideas of the relations between the spectrum of an element and the masses of its atoms. The new departure was supported by Soddy [1] but received some adverse criticism on the ground of insufficient evidence. The later work bearing on this point will be described in Chapter X.

Already in 1911 the theory of the " Nucleus Atom " [2] had been formulated. This gave the first hint as to the physical meaning of chemical and spectroscopic identity, namely that the nuclei of atoms might vary in their mass but yet, at the same time, possess some property in common with each other, namely nuclear charge, upon which the chemistry and spectra depend.

In 1912 appeared the electrochemical work of Hevesy,[3]

[1] Soddy, *Chem. News*, Feb. 28, 1913.     [2] *V.* p. 92.
[3] G. Hevesy, *Phil. Mag.*, **23**, 628,  1912 ; *Physikal. Zeitsch.* **15**, 672, 715,  1912.

which led to the discovery of the remarkable field of research opened up by the use of radioactive bodies as indicators.[1]    A little later Paneth and Hevesy were able to show the complete identity of the electrochemical properties of Radium D and Lead.[2]

In 1914 Rutherford and Andrade [3] examined the self-excited X-ray spectrum of radium B. They used a crystal of rock salt for the analysis and got rid of the effect of the swift $\beta$ rays by putting the source in a strong magnetic field. The wave length of the L radiation proved to be exactly that expected for lead from Moseley's experiment.[4] This was the first proof that isotopes had identical X-ray spectra. The actual values for ordinary lead were subsequently determined by Siegbahn and found to be in excellent agreement with Rutherford and Andrade's results.

**7. The Chemical Law of Radioactive change.**—This law, put in the briefest form, asserts : A radioactive element when it loses an alpha particle goes back two places in the Periodic Table ; when it loses a beta particle it goes forward one place.

The law has been associated with the name of Soddy [5] who was the first to suggest, in the form of a valency property, that part of it relating to alpha rays.[6] But in its more complete enunciation, which took place early in 1913, at least four other investigators can claim a share.

Russell was the first to publish a law covering both kinds of rays,[7] but owing to the fact that he failed to realise that the sequence of elements in the periodic table is a continuous expression, his statement was not so simple and definite as it might have been. Fajans,[8] using as foundation the electrochemical

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[1] *V.* p. 19.

[2] Paneth and Hevesy, *Sitzungber. K. Akad. Wiss. Wien*, iiA, **123,** 1037,  1913.

[3] Rutherford and Andrade, *Phil. Mag.* **27,** 854, 1914.    [4] *V.* p. 93.

[5] *V.* Stewart, *Recent Advances in Physical and Inorganic Chemistry*, Longmans, 1919.

[6] Soddy, *The Chemistry of the Radio Elements*, 29, First Edition, Longmans,  1911.

[7] Russell, *Chem. News*, Jan. 31, 1913.

[8] Fajans, *Physikal. Zeitsch.* Feb. 15, 1913.

results of Hevesy,[1] and Soddy, working on the results of a very full chemical investigation carried out at his request by Fleck,[2] published the generalisation in its full and complete form independently, and practically at the same time.

This law, which will be shown later to be a natural consequence of the much wider generalisation discovered by Moseley,[3] has been of the greatest value in correlating the numerous products of radioactive change, and predicting with accuracy which of them will have identical properties. To the latter the name Isotopes was applied by Soddy in the following words : " The same algebraic sum of the positive and negative charges in the nucleus when the arithmetical sum is different gives what I call ' isotopes ' or ' isotopic elements ' because they occupy the same place in the periodic table. They are chemically identical, and save only as regards the relatively few physical properties which depend upon atomic mass directly, physically identical also." Any element which is the result of a series of changes involving the loss of twice as many beta particles as alpha particles must clearly be the isotope of the parent element, since it must inevitably, by the above law, reach the same place in the periodic table at the end of these operations.

**8. Isobares.**—Just as we can have elements of the same chemical properties but different atomic weight so we can also have those with the same atomic weight but different chemical properties. These Stewart[4] has called " Isobares." Any product due to the loss of a beta ray (which has a negligible mass)[5] must be an isobare of its parent substance, for, without change of mass, it has moved in the periodic table and so changed its chemical properties. It is interesting to note in this connection that no isobare has actually been discovered among the non-radioactive elements as yet, but they must certainly exist.[6]

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[1] Hevesy, *Physikal. Zeitsch.* Jan. 15, 1913.
[2] Fleck, *Trans. Chem. Soc.* **103**, 381, 1052, 1913.
[3] *V.* p. 93.
[4] Stewart, *Phil. Mag.* **36**, 326, 1918.
[5] *V.* p. 91.
[6] *V.* p. 77.

**9. The Radioactive Transformations.**—The radioactive elements are all formed from the two parent elements uranium and thorium by a series of changes or transformations. These changes can be classified according to their nature into two types. In the first type of change called the $\alpha$ ray change the atom loses a particle of mass 4 carrying two positive charges $(+2e)$ which has been identified with the nucleus of the helium atom.[1] In the second or $\beta$ ray change the particle shot off has a negligible mass and carries a single negative charge $(-e)$. Hence in an $\alpha$ ray change the element loses 4 units in atomic weight, while in a $\beta$ ray change its weight is unaltered.

The rate of decay of an element is measured by the " half value " period which may vary from $10^{10}$ years to $10^{-11}$ of a second. The velocity with which the rays are ejected also varies and is apparently connected with the period of the element by the very interesting relation of Geiger and Nuttall.[2] The intricate researches by which the complex series of transformations have been explained belong to the subject of Radioactivity and cannot be described here. From the point of view of isotopes it will be enough to consider the final results which are given in the two diagrams (Figs. 1 and 2).

In the first of these, which is due to Soddy,[3] the nuclear charge or Atomic number,[4] upon which all the chemical and spectroscopic properties of the elements depend, and which expresses its position in the periodic table, is indicated by a series of columns edged with thick lines sloping downwards to the right. The atomic weights are shown by fine lines sloping in the opposite direction. The lines corresponding to even atomic weights have been omitted to simplify the scheme. All elements lying in the same column will therefore be *isotopes* and all elements lying on the same line sloping up to the right will be *isobares*. The $\alpha$ and $\beta$ ray changes are shown by arrows and the period of decay of the elements indicated by times expressed in suitable units.

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[1] $V$. Table p. 106.

[2] Rutherford, *Radioactive Substances and their Radiations*, p. 607, Cambridge, 1913.

[3] Soddy, *Trans. Chem. Soc.*, **115**, 16, 1919.     [4] $V$. p. 93.
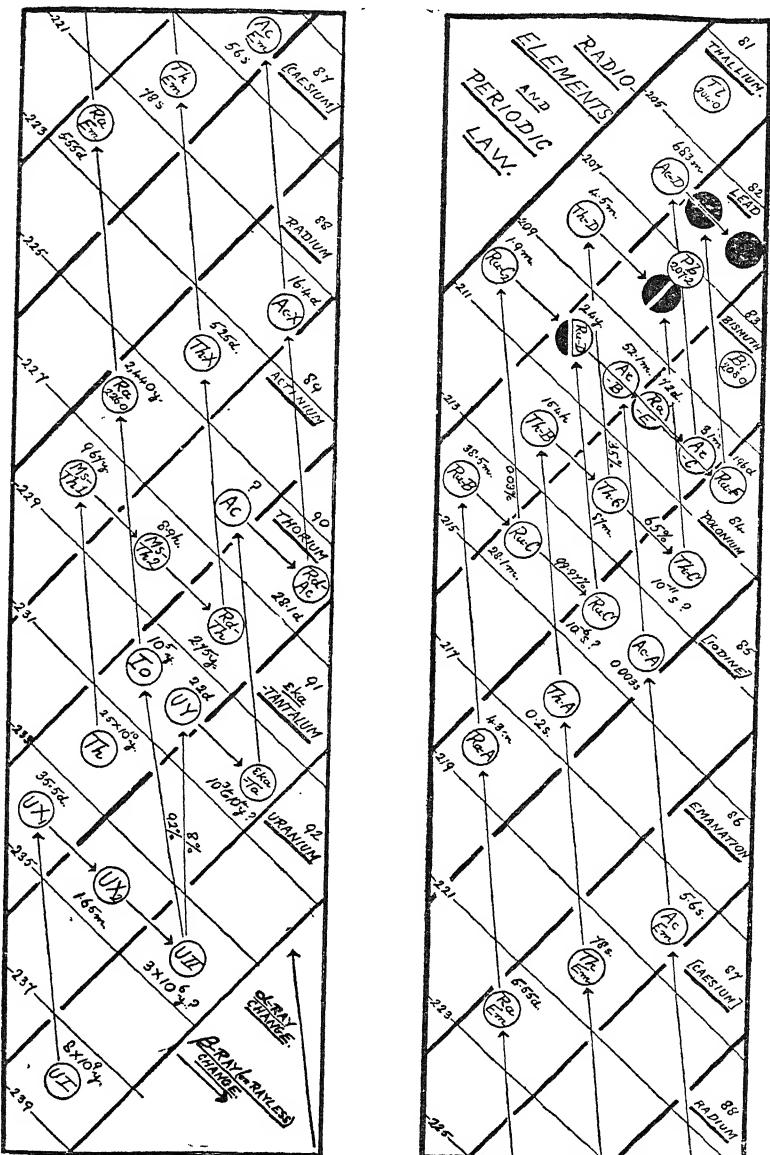
Fig. 1.—Diagram of the transformations of the radio-elements showing atomic number, atomic weight and period of disintegration of each product.

The second diagram [1] is arranged in a simple manner to show the general chains of transformation at a glance. In it the $\alpha$ and $\beta$ ray changes are plotted against atomic number and the other information omitted. On this diagram all elements lying on the same horizontal level will be isotopes. To take an example, uranium I which has an atomic number 92 and an atomic weight 238 loses one $\alpha$ particle and becomes uranium X, atomic number 90, atomic weight 234. This then gives off two $\beta$ rays in succession, first becoming uranium $X_2$
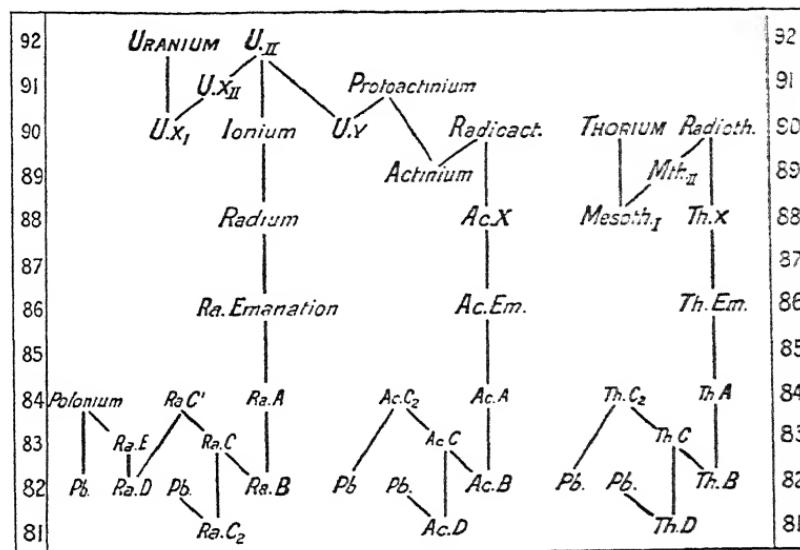


Fig. 2.—Diagram of the radioactive transformations in relation to atomic numbers. In every case a step two downwards is accompanied by the emission of an $\alpha$ particle and one downwards by a $\beta$ particle.

and then Uranium II. Uranium II has an atomic number 92 so that it is an isotope of uranium I. It has an atomic weight 234 so it is an isobare of uranium $X_1$ and uranium $X_2$. Uranium II can disintegrate by shooting off an $\alpha$ particle in two different ways ; about 8 per cent. of its atoms appear to form uranium Y, which is probably the parent substance of the actinium series. Disregarding this for the moment and following the main chain, 92 per cent. of the atoms of uranium

II suffer an $\alpha$ ray change and are transformed into ionium, atomic weight 230, atomic number 90. Ionium loses an $\alpha$ particle and becomes radium, atomic weight 226. This by the same process changes to radium emanation, then to radium A, and then to radium B with atomic weight 214. We see that uranium II has lost 5 $\alpha$ particles in succession, thereby coming back 10 places, 92–82 in the periodic table, and its atomic weight has been reduced 20 units in the process. Radium B loses a $\beta$ particle, becoming radium C which can disintegrate in two different ways. An extremely small proportion, 0·03 per cent., of its atoms undergo an $\alpha$ ray change to radium $C_2$ which then loses a $\beta$ particle and may become inactive lead of atomic weight 210. The vast majority of the atoms of radium C lose a $\beta$ particle and form radium C'. This next loses an $\alpha$ particle and becomes radium D, an active isotope of lead of atomic weight 210. Radium D now loses two $\beta$ particles in succession, becoming radium E and then radium F, which is also called polonium. This finally undergoes its last $\alpha$ ray change and becomes inactive uranium lead of atomic weight 206.

The thorium and actinium chains can be followed on the diagrams in the same manner, but in the case of actinium the parent elements are not satisfactorily settled so that the atomic weights in this series are all doubtful.

**10. The Atomic Weight of Lead.**—The theory of Isotopes of which Professor Soddy had proved himself so prominent an advocate and defender, received its most triumphant vindication, as far as it concerned the products of radioactivity, at the hands of the very chemists who had most reason to doubt its general application, the specialists in the determination of atomic weights.

The charts of radioactive disintegration [1] show that the final product of every series is lead. If we take the main chain of the uranium-radium transformation this lead must have an atomic weight 206, for it has lost 5 alpha particles—each of weight 4—since it was radium, and the atomic weight of radium is 226. On the other hand if we take the main thorium chain

the lead end product must be 6 alpha particles lighter than thorium (232·15) and so should have an atomic weight about 208.

Now ordinary lead, from non-radioactive sources has an atomic weight 207·20, so Soddy [1] suggested in 1913 that the lead derived from minerals containing uranium but no thorium might have a smaller atomic weight than ordinary lead, and on the other hand the atomic weight of lead from minerals containing thorium but no uranium might be greater.

The first experiments were made by Soddy and Hyman [2] with a very small quantity of lead from Ceylon Thorite. This gave a perceptibly higher atomic weight than ordinary lead. Later a large quantity of the same mineral was available. The lead from this when carefully purified gave a density 0·26 per cent. higher than that of common lead. On the assumption that the atomic volumes of isotopes are equal this figure corresponds to an atomic weight of 207·74. A chemical atomic weight determination gave 207·694. A sample of the same lead was sent to Vienna where Professor Honigschmid, a well known expert in such matters, obtained from it a value 207·77 as a mean of eight determinations. These figures not only showed that thorium lead had a higher atomic weight than ordinary lead but also that their atomic volumes were identical, as expected from theory.[3]

At the same time as this work was in progress, the leading American authority on atomic weights, T. W. Richards of Harvard, started a series of investigations on lead derived from various radioactive minerals.[4] The samples of lead from uranium minerals all gave results lower than ordinary lead, as was expected, and one particularly pure specimen of uranio-lead from Norwegian cleveite gave 206·08,[5] a very striking agreement with theory. The following table of properties is taken from his Presidential address to the American Association at Baltimore, December, 1918.

[1] Soddy, *Ann. Rep. Chem. Soc.*, 269, 1913.
[2] Soddy and Hyman, *Trans. Chem. Soc.*, **105**, 1402, 1914.
[3] Soddy, *Roy. Ins.*, May 18, 1917.
[4] Richards and Lembert, *J. Amer. Chem. Soc.*, **36**, 1329, 1914.
[5] Richards and Wadsworth, *J. Amer. Chem. Soc.*, **38**, 2613, 1916.

| | Common Lead. | Mixture Australian. | Uranio-Lead. | Percentage Difference. | |
|---|---|---|---|---|---|
| | A | B | C | A–B | A–C |
| Atomic weight . . . . | 207·19 | 206·34 | 206·08 | 0·42 | 0·54 |
| Density. . . . . . | 11·337 | 11·280 | 11·273 | 0·42 | 0·56 |
| Atomic volume . . . . | 18·277 | 18·278 | 18·281 | 0·01 | 0·02 |
| Melting point (absolute) . . | 600·53 | 600·59 | — | 0·01 | — |
| Solubility (of nitrate) . . | 37·281 | 37·130 | — | 0·41 | — |
| Refractive Index (nitrate) . | 1·7815 | 1·7814 | — | 0·01 | — |
| Thermoelectric effect. . . | — | — | — | 0·00 | — |
| Spectrum wave-length . . | — | — | — | 0·00 | 0·00 |

In further confirmation Maurice Curie in Paris [1] reported 206·36 for a lead from carnotite, and a still lower figure, 206·046, was obtained by Honigschmid in Vienna for a lead from the very pure crystallised pitchblende from Morogoro. This is the lowest atomic weight found so far. The highest, 207·9, was also determined by Honigschmid for lead from Norwegian thorite.[2]

**11. Atomic weights of Thorium and Ionium.**—Although the above results obtained with lead are far the most conclusive and important it is not the only element which affords direct experimental evidence of the different atomic weights of isotopes. The atomic weight of ionium, calculated by adding the weight of one alpha particle to the atomic weight of its product, radium, is 230, whereas that of thorium, its isotope, is slightly above 232. Joachimsthal pitchblende contains hardly any thorium so that an ionium-thorium preparation separated by Auer von Welsbach from 30 tons of this mineral might be regarded as containing a maximum concentration of ionium. On the other hand the period of thorium is about $10^5$ times longer than that of ionium so that it was doubtful if even in this preparation there would be enough ionium to show a difference in atomic weight. Honigschmid and Mlle. Horovitz have made a special examination of this point, first redetermining as accurately as possible the atomic weight of thorium and then that of the thorium-ionium prepar-

[1] M. Curie, *Compt. Rend.*, **158**, 1676, 1914.
[2] Honigschmid, *Zeit. Elektrochem.*, **24**, 163, 1918 ; **25**, 91, 1919.

ation from pitchblende. They found 232·12 for the atomic
weight of thorium, and by the same careful method 231·51
for that of the thorium-ionium.

**12. Use of radioactive isotopes as indicators.**—Con-
sider an inactive element A which has a radioactive isotope B.
If these are mixed together in any proportions no chemical
or physical process known is capable of altering the ratio of
the proportions of this mixture to any measurable extent.
Now the radioactive methods of detecting and measuring B
are many millions of millions of times more delicate than
the chemical methods of detecting and measuring A, so that
by mixing with A a small quantity of B we can trace its presence
far beyond the limits of chemical analysis. We have, as it
were, marked the atoms of A with an indelible label so that
the minutest trace of the element can be measured with ease
and certainty.

By this powerful and novel device, which has been developed
by G. Hevesy [1] $10^{-9}$ gr. of lead can be determined quantita-
tively and solution concentrations can be dealt with down to
$10^{-14}$ of normal. By adding radium D to the lead salt and
estimating it electroscopically the solubility of lead sulphide
and chromate, and the amount of lead chloride carried down
in a silver chloride precipitate, may readily be determined.

Recently, by the same principle, it has been shown that a
free exchange of the metallic atom among the competing acid
radicles occurs for ionised, but not for non-ionised, compounds.
The general method was to mix solutions of two different
compounds of lead in equimolecular proportions, the one
compound only being " activated " by presence of thorium-B
(which is isotopic with lead), and to determine the activity of
the lead in the less soluble compound crystallising out. When
active lead nitrate and inactive lead chloride are dissolved
in molecular proportion in boiling pyridine, the lead in the
lead chloride crystallising out is half as active as the lead in
the original lead nitrate, but when such an active lead salt
is so mixed with an organic compound of lead, such as lead
tetraphenyl or diphenyl nitrate, in suitable solvents, no inter-

[1] Hevesy, Brit. Assoc., 1913 ; *Chem. News*, Oct. 13, 166,  1913.

change of lead occurs, and the active lead salt retains its original activity. This constitutes something like a direct proof of the ionic dissociation theory and of the current views as to the difference between the nature of chemical union in electrolytes and non-electrolytes. When the acetates of quadrivalent activated lead and of bivalent inactive lead are mixed in glacial acetic acid, the activity of the first compound, after crystallising out from the mixture, is reduced to one half. This indicates, since the two lead ions differ only by two electrons, a free interchange of electrons between them and a dynamic equilibrium between ions and electrons and between free electrons and the electrodes in electrolysis.[1]

Isotopes can also be used to determine the velocity of diffusion of molecules among themselves.[2] The rate of diffusion is dependent on the molecular diameter, and not on the mass, so that a radioactive element diffusing among the inactive molecules of its isotope affords a means of investigating this otherwise insoluble problem. The experiment has been tried with molten lead. At the bottom of a narrow vertical tube was placed a layer of lead rendered active by the presence of thorium-B, and above it a layer three times the height of common lead. The whole was kept at 340° for several days. After cooling, the cylinder was cut into four equal lengths, each melted and hammered into foil, and the concentration of thorium-B in each determined by alpha ray measurements. Values for the diffusion coefficient between 1·77 and 2·54 per sq. cm. per day, with a mean of 2·22 in seventeen experiments, were obtained. On certain theories of physical chemistry this corresponds with a diameter of the lead molecule between $0·78$ and $1·16 \times 10^{-8}$ cm., according to the formulae used to connect the two quantities. The value found by similar theories when reduced to a temperature of 18° and for a fluid of the viscosity of water, becomes 2·13. Since the value for lead ions diffusing in aqueous solutions is 0·68, this indicates that the molecular diameter in the case

[1] G. Hevesy and L. Zechmeister, *Ber.*, **53B,** 410, 1920; *Zeitsch. Elektrochem.* **26,** 151, 1920.

[2] J. Groh and Hevesy, *Ann. Physik.*, iv., **63,** 85, 1920.

of metallic lead is only a third of that in the case of the ion, and shows that the latter is probably hydrated.[1]

**13. Classification of the radioactive isotopes.**—It is clear that the relations between isotopes formed by radioactive disintegrations need not necessarily have the same simple form as those subsisting between isotopes of the inactive elements. Neuburger,[2] using the nucleus model of the radioelements proposed by Lise Meitner,[3] suggests that the radioactive isotopes may be divided into three or even four classes.

(1) Isotopes of the first class are those which possess only the same nuclear charge and the same arrangement of outer electrons such as radium and mesothorium I.

(2) Isotopes of the second class have, in addition, the same nuclear mass, that is to say the same atomic weight, and the same total number of nuclear " building stones." Examples of this class are ionium and uranium Y.

(3) Isotopes of the third class still possess the same number of each nuclear building stone, but they have a different arrangement of these in the atomic nucleus, and thus possess different chances of disintegrating, such as Radium D and Actinium B.

(4) Isotopes of the fourth class would be those possessing the same arrangement of nuclear building stones in the atomic nucleus, and thus the same probability of disintegrating. Such isotopes actually exist, but we have no available means of distinguishing between them. Hence we cannot at present designate them definitely as isotopes. Examples of these are radium $C_2$ and actinium D.

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[1] Soddy, *Ann. Rep. Chem. Soc.*, 227, 1920.

[2] Neuburger, *Nature*, **108**, 180, 1921.

[3] Meitner, *Die Naturwissenschaften*, **9**, 423, 1921.